10/018847
JC10 Rec'd PCT/PTO 18 DEC 2001

Atty. Docket #: 1999/G-014

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INTERNATIONAL APPL. NO.:	PCT/EP00/05759	:	
INTERNATIONAL FILING DATE:	-JUNE 21, 2000-	:	
APPLICANT:	FRANK OSAN ET AL	:	
SERIAL NO:	TO BE ASSIGNED.	:	ART UNIT:
FILED:	-HEREWITH-	:	EXAMINER:
FOR:	"METHOD OF PRODUCING AMORPHOUS POLYOLEFINS WITH A WIDE MOLE WEIGHT DISTRIBUTION"	:	

Commissioner for Patents
Box PCT
P.O. Box 2327
Arlington, VA 22202

"Express Mail" No.: ET284672057 Date: -DECEMBER 18, 2001-

I hereby certify that this paper, along with any other paper or fee referred to in this paper as being transmitted herewith, is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10, postage prepaid, on the date indicated above, addressed to the Commissioner for Patents, P.O. Box 2327, Arlington, VA 22202.

-Carrie A. McPherson-
(Typed or printed name of mailing paper or fee)

Carrie A. McPherson
(Signature of person mailing paper)

TRANSMITTAL OF APPLICATION PAPERS TO U.S. DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. §371 (37 CFR 1.494 OR 1.495)

This Transmittal Letter is based upon PTO Form 1390 (as revised in May, 1993).

The above-identified applicant(s) (jointly with their assignee) have filed an International Application under the P.C.T. and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information.

1. [X] This is a FIRST submission of items concerning a filing under 35 U.S.C. §371.

2. [ ] This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. §371.

3. [X] This is an express request to begin national examination procedures (35 U.S.C. §371[f]) at any time rather than delay.

4. [X] A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) within the time period required.

5. [X] A copy of the International Application as filed (35 U.S.C. §371[c][2]) --
   a. [X] is transmitted herewith (required when not transmitted by International Bureau).
   b. [ ] has been transmitted by the International Bureau. See WIPO Publication WO 01/02481.
   c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).

6. [X] A (verified) translation of the International Application into the English language is enclosed -with- Two (2) Sheets of Drawings.

7. [ ] Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. 371[c][3])
   a. [ ] are transmitted herewith (required if not transmitted by the International Bureau).
   b. [ ] have been transmitted by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [ ] have not been made and will not be made.
   e. [ ] will be submitted with the appropriate surcharge.

8. [ ] A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §371[c][3]) is enclosed or will be submitted with the appropriate surcharge.

9. [X] An oath or declaration/power of attorney of the inventor(s) (35 U.S.C. §371[c][4]) will follow.
   [ ] and is attached to the translation of (or a copy of) the International Application.
   [ ] and is attached to the substitute specification.

10. [ ] A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371[c][5]) is enclosed.

Items 11. to 16. below concern other document(s) or information included:

11. [X] An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.

12. [X] An Assignment for recording and a separate cover sheet in compliance with 37 CFR 3.28 and 3.31 will follow.

13. [X] A FIRST preliminary amendment is enclosed.
    A SECOND or SUBSEQUENT preliminary amendment is enclosed.

14. [ ] A substitute specification (including claims, abstract, drawing) is enclosed.

15. [ ] A change of power of attorney and/or address letter is enclosed.

16. [X] Other items of information:

[X] This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any missing parts will be filed before expiration of--

[ ] 22 months from the priority date under 37 CFR 1.494(c), or

[X] 32 months from the priority date under 37 CFR 1.495(c).

[X] The undersigned attorney is authorized by the International applicant and by the inventors to enter the National Phase pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

International Application No. PCT/EP00/05759 1999/G-014

[X] Receiving Office: EPO
[X] IPEA (if filing under 37 CFR 1.495): EPO
[X] Priority Claim(s) (35 USC §§ 119, 365):
German Appln. 199 29 809.2 filed -June 30, 1999-.
[X] A copy of the International Search Report is

[ ] enclosed.
[x] attached to the copy of the International Application.

[X] A copy of the Receiving Office Request Form is enclosed.
[X] Form PTO/SB/05 (1) sheet
[X] Form PCT/IB/308 (1) sheet
[X] Form PCT/RO/101 (4) pages
[X] AMENDED SHEETS pp. 22-26 IN ENGLISH (claims 1-12)
[X] AMENDED SHEETS pp. 20-24 IN GERMAN (claims 1-12)

The fee calculation is set forth on the next page of this Transmittal Letter.

FEE CALCULATION SHEET

[x] A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

Basic Fee....................................	$ 890.00
Total Number of claims in excess of (20) times $18..................	-0-
Number of independent claims in excess of (3) times $84.................	-0-
Fee for multiple dependent claims $280................................	-0-
TOTAL FILING FEE...	$ 890.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue upon allowance is authorized. An additional copy of this page is attached.

Respectfully submitted,

By Ashley I. Pezzner

Ashley I. Pezzner
Reg. No. 35,646
CONNOLLY BOVE LODGE & HUTZ LLP
1220 Market Street
P.O. Box 2207
Wilmington, Delaware 19899
Tel. (302) 658-9141

AIP/cam (5587*324)
Enclosures
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FEE CALCULATION SHEET

[x] A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492).

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Total Number of claims in excess of (20) times $18....................	-0-
Number of independent claims in excess of (3) times $84.................	-0-
Fee for multiple dependent claims $280..................................	-0-
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Respectfully submitted,

By Ashley I. Pezzner

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AIP/cam (5587*324)
Enclosures
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10/018847
JC13 Rec'd PCT/PTO 18 DEC 2001

Please type a plus sign (+) inside this box → [+]

PTO/SB/05 (4/98)
Approved for use through 09/30/2000. OMB 0651-0032
Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 C.F.R. § 1.53(b))

Attorney Docket No.	1999/G-014
First Inventor or Application Identifier	FRANK OSAN ET AL
Title	METHOD OF PRODUCING AMORPHOUS...
Express Mail Label No.	ET284672057

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

ADDRESS TO: Assistant Commissioner for Patents
Box Patent Application
Washington, DC 20231

1. [X] * Fee Transmittal Form (e.g., PTO/SB/17) (Submit an original and a duplicate for fee processing)
2. [X] Specification [Total Pages 27] (preferred arrangement set forth below)
   - Descriptive title of the Invention
   - Cross References to Related Applications
   - Statement Regarding Fed sponsored R & D
   - Reference to Microfiche Appendix
   - Background of the Invention
   - Brief Summary of the Invention
   - Brief Description of the Drawings (if filed)
   - Detailed Description
   - Claim(s)
   - Abstract of the Disclosure
3. [X] Drawing(s) (35 U.S.C. 113) [Total Sheets 2]
4. Oath or Declaration [Total Pages]
   a. [ ] Newly executed (original or copy)
   b. [ ] Copy from a prior application (37 C.F.R. § 1.63(d)) (for continuation/divisional with Box 16 completed)
      i. [ ] DELETION OF INVENTOR(S) Signed statement attached deleting inventor(s) named in the prior application, see 37 C.F.R. §§ 1.63(d)(2) and 1.33(b).

*NOTE FOR ITEMS 1 & 13: IN ORDER TO BE ENTITLED TO PAY SMALL ENTITY FEES, A SMALL ENTITY STATEMENT IS REQUIRED (37 C.F.R. § 1.27), EXCEPT IF ONE FILED IN A PRIOR APPLICATION IS RELIED UPON (37 C.F.R. § 1.28).

5. [ ] Microfiche Computer Program (Appendix)
6. Nucleotide and/or Amino Acid Sequence Submission (if applicable, all necessary)
   a. [ ] Computer Readable Copy
   b. [ ] Paper Copy (identical to computer copy)
   c. [ ] Statement verifying identity of above copies

ACCOMPANYING APPLICATION PARTS

7. [ ] Assignment Papers (cover sheet & document(s))
8. [ ] 37 C.F.R.§3.73(b) Statement (when there is an assignee) [ ] Power of Attorney
9. [X] English Translation Document (if applicable)
10. [X] Information Disclosure Statement (IDS)/PTO-1449 [X] Copies of IDS Citations
11. [X] Preliminary Amendment
12. [X] Return Receipt Postcard (MPEP 503) (Should be specifically itemized)
13. [ ] * Small Entity Statement(s) (PTO/SB/09-12) [ ] Statement filed in prior application, Status still proper and desired
14. [ ] Certified Copy of Priority Document(s) (if foreign priority is claimed)
15. [X] Other: Form PCT/IB/308 (1) sheet
16 X AMENDED SHEETS pp22-26 ENG
17 X AMENDED SHEETS pp20-24 GERMAN
18 X Receiving Office Request Form PCT/RO/101 (4) pages

16. If a CONTINUING APPLICATION, check appropriate box, and supply the requisite information below and in a preliminary amendment:
[ ] Continuation [ ] Divisional [ ] Continuation-in-part (CIP) of prior application No: ______/______
Prior application information: Examiner ______ Group / Art Unit: ______

For CONTINUATION or DIVISIONAL APPS only: The entire disclosure of the prior application, from which an oath or declaration is supplied under Box 4b, is considered a part of the disclosure of the accompanying continuation or divisional application and is hereby incorporated by reference. The incorporation can only be relied upon when a portion has been inadvertently omitted from the submitted application parts.

17. CORRESPONDENCE ADDRESS

[ ] Customer Number or Bar Code Label (Insert Customer No. or Attach bar code label here) or [X] Correspondence address below

Name	Ashley I. Pezzner, Esquire				
	CONNOLLY BOVE LODGE & HUTZ LLP				
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	P.O. Box 2207				
City	Wilmington	State	DE	Zip Code	19899
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Name (Print/Type)	ASHLEY I. PEZZNER	Registration No. (Attorney/Agent)	35,646
Signature	Ashley I. Pezzner	Date	12/18/2001

Burden Hour Statement: This form is estimated to take 0.2 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Box Patent Application, Washington, DC 20231.

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JC13 Rec'd PCT/PTO 18 DEC 2001

ATTORNEY DOCKET NO.: 1999/G 014 (5587*324)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT: FRANK OSAN *ET AL.*	)
SERIAL NO. TO BE ASSIGNED	) ART UNIT: TO BE ASSIGNED
FILED: HEREWITH	) EXAMINER: TO BE ASSIGNED
FOR: METHOD OF PRODUCING AMORPHOUS POLYOLEFINS WITH A WIDE MOLE WEIGHT DISTRIBUTION	)

Asst. Commissioner for Patents
Washington, D.C. 20231

"EXPRESS MAIL" NO. ET 284 672 057 DATE: DECEMBER 18, 2001

I HEREBY CERTIFY THAT THIS PAPER OR FEE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE "EXPRESS MAIL POST OFFICE TO ADDRESSEE" SERVICE UNDER 37 CFR 1.10 ON THE DATE INDICATED AND IS ADDRESSED TO THE U.S. PATENT AND TRADEMARK OFFICE, P.O. BOX 2327, ARLINGTON, VA 22202

CARRIE A. MCPHERSON
(TYPED OR PRINTED NAME OF PERSON MAILING PAPER OR FEE)

Carrie A. McPherson
(SIGNATURE OF PERSON MAILING PAPER OR FEE)

10018047 .122701

PRELIMINARY AMENDMENT

Sir:

Prior to fee calculation and examination please amend the above-identified application as follows.

In the Claims

Please cancel claims 1-10.

Please add the following new claims.

--11. A process for the continuous preparation of a bimodal or multimodal mixture of two or more amorphous polyolefins having a different molar mass, wherein the viscosity ratio of at least two amorphous polyolefins having a different molar mass is less than 0.005 and greater than 4 and a) the amorphous polyolefin having a high molar mass is prepared by

solution polymerization in one reactor of an assembly of two or more reactors connected in parallel or in series and the other constituents of the mixture are produced in the other reactors after which the polyolefins are mixed in solution, or b) the amorphous polyolefin having a high molar mass is prepared by solution polymerization in one reactor and the other constituents of the mixture are introduced in the form of a polymer solution into the solution flowing from the reactor, and the solution of polymer mixture obtained according to a) or b) is homogenized and the solvent is separated off.

12. The process as claimed in claim 11, wherein the amorphous polyolefin having a high molar mass has a VN of > 80 ml/g and an M_w of > 90,000 g/mol.

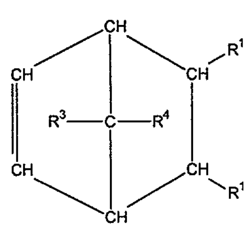
13. The process as claimed in claim 11, wherein the amorphous polyolefin having a high molar mass has a VN of > 100 ml/g and an M_w of > 100,00 g/mol.

14. The process as claimed in claim 11, wherein the amorphous polyolefin having a high molar mass has a VN of > 120 ml/g and an M_w of > 120,000 g/mol.

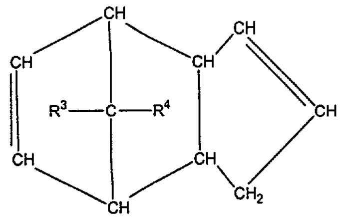
15. The process as claimed in claim 11, wherein the amorphous polyolefin having a high molar mass has a VN of >150 ml/g and an M_w of > 150,000 g/mol.

16. The process as claimed in claim 11, wherein the amorphous polyolefin is a cycloolefin copolymer.

17. The process as claimed in claim 11, wherein the bimodal or multimodal mixture comprises at least one cycloolefin copolymer comprising from 0.1 to 100% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from at least one polycyclic olefin of the formula I, II, II', III, IV, V or VI.



(I),



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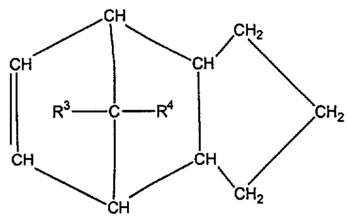

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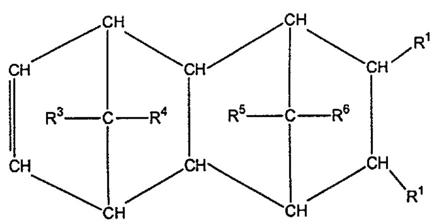

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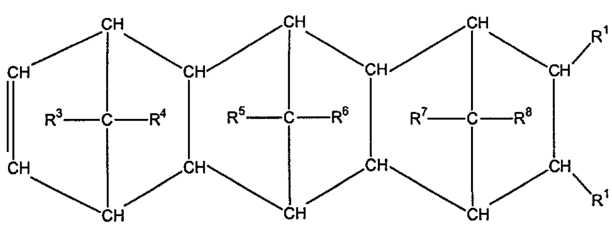

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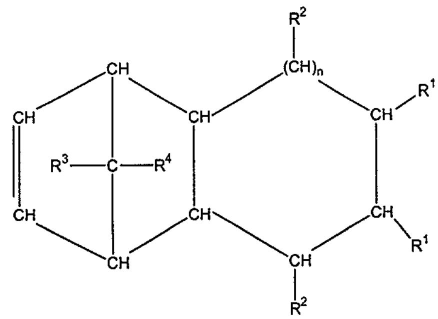

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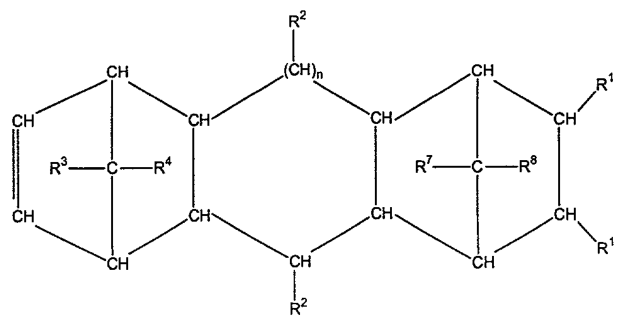
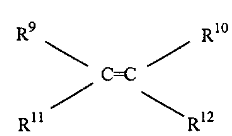


(VI),

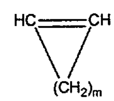
wherein R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and are each a hydrogen atom or a C_1-C_{20}-hydrocarbon radical, or form a saturated, unsaturated or aromatic ring, wherein identical radicals R^1 to R^8 in the various formulae 1 to VI can have different meanings, and n is from 0 to 5, and, optionally, up to 99.9% by weight, based on the total mass of the cycloolefin polymer, of polymerized units derived from one or more acyclic olefins of the formula VII



(VII),

wherein R^9, R^{10}, R^{11} and R^{12} are identical or different and are each a hydrogen atom, a linear, branched, saturated or unsaturated C_1-C_{20}-hydrocarbon radical.

18. The process as claimed in claim 17, wherein the cycloolefin copolymers further comprise up to 45% by weight, based on the total mass of the cycloolefin copolymer, or polymerized units derived from one or more monocyclic olefins of the formula VIII



(VIII)

wherein m is from 2 to 10.

19. The process as claimed in claim 18, wherein the cyclic and polycyclic olefins contain one or more substituents selected from the group consisting of halogen, hydroxyl, ester, alkoxy, carboxy, cyano, amido, imido and silyl.

20. The process as claimed in claim 18, wherein the cycloolefin copolymers comprise polymerized units derived from polycyclic olefins of the formula I or III and polymerized units derived from acyclic olefins of the formula VII.

21. The process as claimed in claim 18, wherein the cycloolefin copolymers comprises polymerized units derived from olefins having a norbornene skeleton.

22. The process as claimed in claim 18, wherein the cycloolefin copolymers comprise polymerized units derived from acyclic α-olefins having from 2 to 20 carbon atoms.

23. The process according to claim 18, wherein R^9, R^{10}, R^{11} and R^{12} are identical or different and are each a hydrogen atom, a C_1-C_8 alkyl radical or a C_6-C_{18} aryl radical.

24. The process as claimed in claim 21, wherein the cycloolefin copolymer comprise norbornene, tetracyclododecene, vinylnorbornene or norbornadiene.

25. The process as claimed in claim 22, wherein the α-olefin is ethylene.

26. The process according to claim 22, wherein the α-olefin is propylene.

27. The process according to claim 18, wherein R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and are each a hydrogen atom, a C_1-C_8 alkyl radical, C_6-C_{18} aryl radical, a C_7-

C_{20} alkylenearyl radical, a cyclic or acyclic C_2-C_{20} alkenyl radical or form a saturated, unsaturated or aromatic ring.- -

REMARKS

The applicants respectfully request that the preliminary amendment be entered prior to fee calculation and examination. Support for newly added claims 11-27 can be found in the amended sheets of claims 1-12. In addition, support can be found in the specification at pages 1 and 12. The claims have been amended similar to the PCT (see the amended sheets of the PCT). No additional fee is required for the extra claims. If there are any additional fees due in connection with the filing of this response, the Commissioner is authorized to charge or credit any overpayment to Deposit Account No. 03-2775.

A prompt and favorable action is solicited.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

By____________________
Ashley I. Pezzner
Reg. No. 35,646
Tel. (302) 888-6270

AIP/cam
178213

METHOD OF PRODUCING AMORPHOUS POLYOLEFINS WITH A WIDE MOLE WEIGHT DISTRIBUTION

Frank Osan
Klaus Berger
Dieter Ruchatz
Oliver Stark
-and-
Toru Nakamura

ENGLISH TRANSLATION
OF
INTERNATIONAL APPLICATION

PCT/EP00/05759 .... IFD: -June 21, 2000-

-with-

Two (2) Sheets of Drawings

1999/G-014 .... (5587*324)

"Express Mail" mailing label number ET284672057
Date of Deposit -December 18, 2001-
I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37CFR 1.10 on the date indicated above and is addressed to Box PCT, Commissioner for Patents, P.O. Box 2327, Arlington, VA 22202
-Carrie A. McPherson-
(Typed or printed name of person mailing paper or fee)
Carrie A. McPherson
(Signature of person mailing paper or fee)

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

WIPO

PCT

(43) International publication date
11 January 2001 (11.01.2001)

(10) International publication number
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(51) International patent classification[7]: C08L 23/04, C08F 232/00

(21) International application number: PCT/EP00/05759

(22) International filing date: 21 June 2000 (21.06.2000)

(25) Language of filing: German

(26) Language of publication: German

(30) Data relating to the priority:
199 29 809.2 30 June 1999 (30.06.1999) DE

(71) Applicant (*for all designated States except US*): TICONA GMBH [DE/DE]; An der B 43, D-65451 Kelsterbach (DE).

(72) Inventors; and
(75) Inventors/Applicants (*US only*): OSAN, Frank [DE/DE]; Hattersheimerstrasse 29, D-65779 Kelkheim (DE). BERGER, Klaus [DE/DE]; Starkeradweg 8, D-65843 Sulzbach (DE). RUCHATZ, Dieter [DE/DE]; Schmalenfeldweg 5a, D-42579 Heiligenhaus (DE). STARK, Oliver [DE/DE]; Dekan-Schuster-Strasse 47, D-63110 Rodgau (DE). NAKAMURA, Toru [JP/JP]; 13-201, Tsukushino 3 chome, Abiko-shi, Chiba 270-1164 (JP).

(81) Designated states (*national*): JP, KR, US.

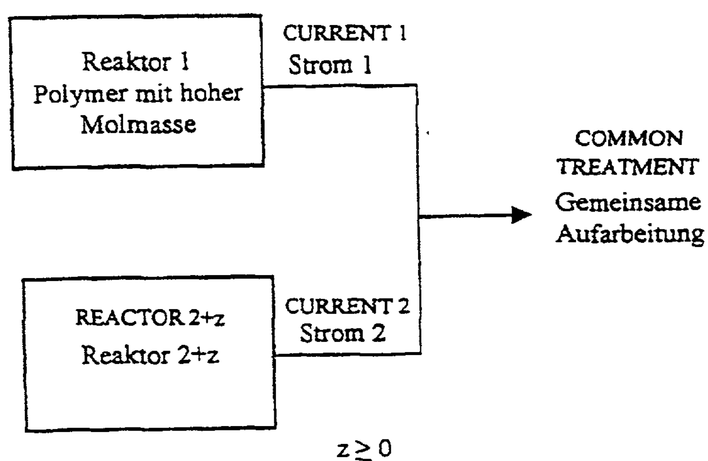
(84) Designated states (*regional*): European Patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

[continued on next page]

As printed

(54) Title: METHOD OF PRODUCING AMORPHOUS POLYOLEFINS WITH A WIDE MOLE WEIGHT DISTRIBUTION

(54) Bezeichnung: VERFAHREN ZUR HERSTELLUNG AMORPHER POLYOLEFINE MIT BREITER MOLMASSENVERTEILUNG



1 REACTOR 1
POLYMER OF HIGH MOLECULAR WEIGHT

(57) Abstract: The invention relates to a method of producing a bimodal or multimodal mixture of amorphous polyolefins of different mole weights. According to the inventive method, at least one amorphous polyolefin of high molecular weight is contacted in a solution with at least one amorphous polyolefin of low molecular weight and is mixed and the solvent is removed.

[continued on next page]

WO 01/02481 A1

Published:
- With the International Search Report.

For an explanation of the two-letter codes and the other abbreviations, reference is made to the explanations ("Guidance Notes on Codes and Abbreviations") at the beginning of each regular edition of the PCT Gazette.

(57) **Zusammenfassung:** Die vorliegende Erfindung betrifft ein Verfahren zur Herstellung eines bimodalen oder multimodalen Gemisches von amorphen Polyolefinen unterschiedlicher Molmasse, wobei mindestens ein amorphes Polyolefin mit hoher Molekularmasse mit mindestens einem amorphen Polyolefin mit niedriger Molekularmasse in Lösung in Kontakt gebracht und gemischt werden und das Lösungsmittel anschließend entfernt wird.

10/018847
JC13 Rec'd PCT/PTO 18 DEC 2001

Description

Process for producing amorphous polyolefins having a broad molar mass distribution.

The invention relates to a continuous process for producing mixtures of amorphous polyolefins having a broad molar mass distribution and a uniform glass transition temperature.

To produce a bimodal or multimodal mixture of amorphous polyolefins, two or more polyolefins having different molar masses have to be mixed and homogenized. In the case of a small difference in the molar masses of the starting components and consequently a small difference in the melt viscosities, mixing can be carried out in the melt. This occurs in the case of extrusion. However, above a certain difference in the melt viscosities, homogeneous mixing of amorphous polyolefins in the melt can no longer be carried out. According to Karam, Bellinger, Ind. A. Chem. Eng. Fund 7(1968) 4, 571-581, this limit is reached when the viscosity ratio of secondary component to main component of the mixture is less than 0.005 and greater than 4. Accordingly, narrow limits are imposed on the effective mixing of a relatively high molecular weight polymer into a low molecular weight matrix via the melt. Melt mixing can only be carried out by means of a number of melt mixers connected in series. However, such a process has high capital costs and process costs and its economics are therefore poor.

EP-A-0 843 223 discloses a bimodal toner. The mixture is produced batchwise. The relatively high molecular weight component has a molecular weight (M_w) of 70,000 g/mol, a viscosity number (VN) of 80 ml/g and a glass transition temperature above 70°C. The toner is produced by mixing the starting components in the melt.

In the case of large differences in the molar masses of the blend components, the melt viscosities differ so much that the production of a homogeneous blend via the melt is possible only with great difficulty.

WO 98/29783 discloses a tower having a broad molar mass distribution (bimodal, multimodal, broad distribution without separate peaks). The preparation of the base material and mixing were carried out batchwise. The relatively high molecular weight component had an M_w of 100 000 g/l and a VN of 130 ml/g, therefore somewhat higher than EP-A-0 483 223. In the case of large differences in the molar mass of the blend components, the melt viscosities differ so greatly the production of a homogeanous blend via the melt is extremley difficult.

EP-A-0 128 045 discloses a process for preparing crystalline polyolefins. The catalyst system for the polymerization of ethylene to form polyethylene comprises two different metallocenes. The process of homogeneous catalysis and the resulting polyethylene having a polydispersity of from 2 to 50 are likewise described. In contrast, corresponding catalyst systems for preparing amorphous cycloolefin polymers are extremely difficult to find. Firstly, they have to catalyze the reaction highly specifically without forming light-scattering by-products which reduce the transparency of the material, and, secondly, these catalysts should display the same copolymerization diagram so that under identical reaction conditions a plastic having only one macroscopically observable glass transition temperature is formed.

WO 96/18662 discloses a process for preparing polyethylene in which the first stage is carried out in a low-boiling hydrocarbon in a loop reactor, the second stage is likewise carried out in a solvent in a loop reactor and the third stage is carried out in the gas

phase. In each stage, further catalyst, cocatalyst, ethylene or hydrogen can be added. The high molecular weight component is prepared in the first stage. In the gas-phase reactor, a C_4-C_8-α-olefin can also be added as comonomer. This process cannot be applied to the preparation of cycloolefin copolymers since gas-phase reactors are unsuitable for liquid comonomers. Furthermore, the catalysts should catalyze the reaction highly specifically without light-scattering by-products being formed.

It is an object of the present invention to provide an economical and environmentally friendly continuous process for producing a bimodal or multimodal mixture of one or more amorphous polyolefins.

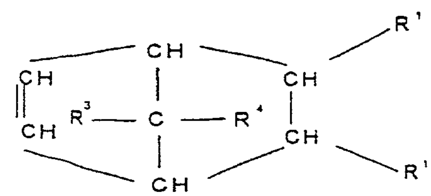
This object is achieved by a process for producing a bimodal or multimodal mixture of amorphous polyolefins regardless of the difference in the mean molar mass and in the molar mass distribution of the amorphous polyolefins, wherein at least one amorphous polyolefin having a high molar mass is brought into contact and mixed with at least one amorphous polyolefin having a low molar mass in solution and the solvent is subsequently removed.

According to the invention, preference is given to a process in which the amorphous polyolefin having a high molar mass has a VN of > 80 ml/g and an M_w of > 90,000 g/mol, preferably a VN of > 120 ml/g and an M_w of > 120,000 g/mol, particularly preferably a VN of > 150 ml/g and an M_w of > 150,000 g/mol. Such polyolefins can be processed only with extreme difficulty in the melt.

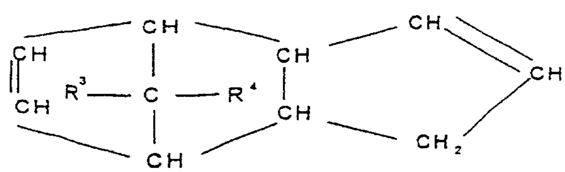
According to the invention, particular preference is given to a process in which the amorphous polyolefin having a high molar mass is a cycloolefin copolymer. Cycloolefin copolymers can readily be prepared

industrially by means of metallocene catalysts or other transition metal compounds.

The bimodal or multimodal mixture comprises at least one cycloolefin copolymer comprising from 0.1 to 100% by weight, preferably from 0.1 to 99.9% by weight, particularly preferably from 10 to 90% by weight and very particularly preferably from 30 to 70% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from at least one polycyclic olefin of the formula I, II, II′, III, IV, V or VI.



(I),



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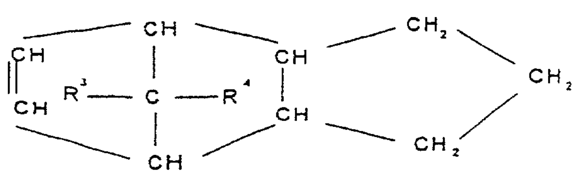

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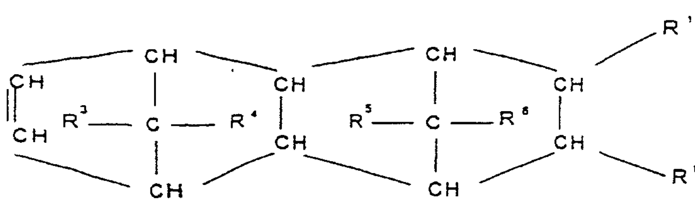
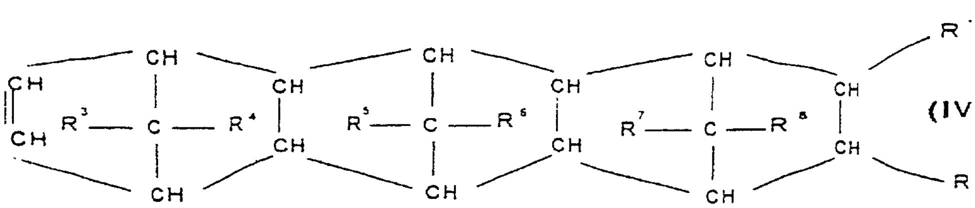
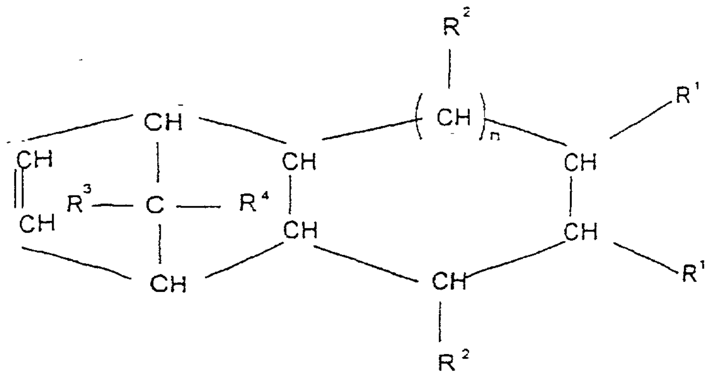


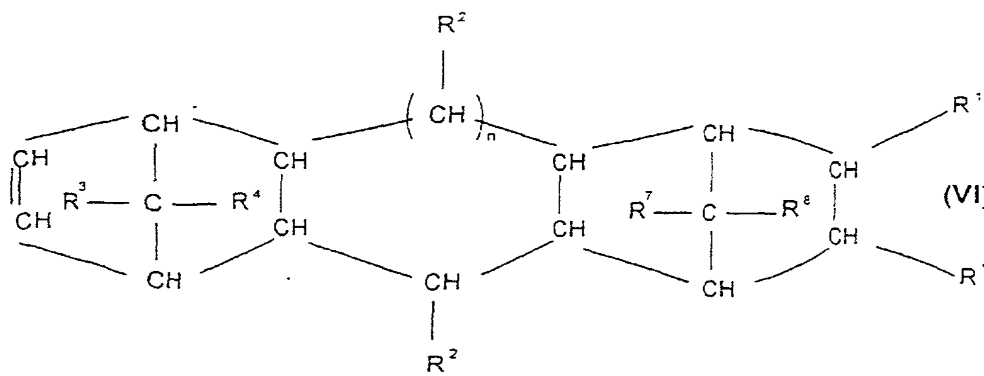
(III),



(IV),

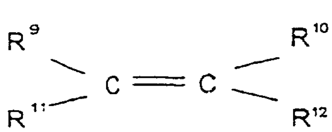


(V),



(VI),

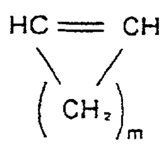
where R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and are each a hydrogen atom or a C_1-C_{20}-hydrocarbon radical such as a linear or branched C_1-C_8-alkyl radical, a C_6-C_{18}-aryl radical, a C_7-C_{20}-alkylenearyl radical or a cyclic or acyclic C_2-C_{20}-alkenyl radical or form a saturated, unsaturated or aromatic ring, where identical radicals R^1 to R^8 in the various formulae I to VI can have different meanings, and n can be from 0 to 5, and, if desired, up to 99.9% by weight, preferably from 0.1 to 99.9% by weight, particularly preferably from 10 to 90% by weight and very particularly preferably from 30 to 70% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from one or more acyclic olefins of the formula VII



(VII),

where R^9, R^{10}, R^{11} and R^{12} are identical or different and are each a hydrogen atom, a linear, branched, saturated or unsaturated C_1-C_{20}-hydrocarbon radical such as a C_1-C_8-alkyl radical or a C_6-C_{18}-aryl radical.

In addition, the cycloolefin copolymers used according to the invention may further comprise from 0 to 45% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from one or more monocyclic olefins of the formula VIII



(VIII),

where m is from 2 to 10.

The cyclic olefins likewise include derivatives of these cyclic olefins containing polar groups such as halogen, hydroxyl, ester, alkoxy, carboxy, cyano, amido, imido or silyl groups.

For the purposes of the invention, preference is given to cycloolefin copolymers which comprise polymerized units derived from polycyclic olefins of the formula I or III and polymerized units derived from acyclic olefins of the formula VII.

Particular preference is given to cycloolefin copolymers which comprise polymerized units derived from olefins having a norbonene skeleton, preferably norbornene, tetracyclododecene, vinylnorbornene or norbornadiene, particularly preferably norbornene or tetracyclododecene.

Particular preference is also given to cycloolefin copolymers which comprise polymerized units derived

from acyclic olefins having terminal double bonds, e.g. α-olefins having from 2 to 20 carbon atoms, very particularly preferably ethylene or propylene. Greatest preference is given to norbornene-ethylene and tetracyclododecene-ethylene copolymers.

It can also be advantageous to use terpolymers, particularly preferably norbornene-vinylnorbornene-ethylene, norbornene-norbornadiene-ethylene, tetracyclododecene-vinylnorbornene-ethylene, tetracyclododecene-vinyltetracyclododecene-ethylene or norbornene-dicyclopentadiene-ethylene terpolymers.

The proportion of polymerized units which are derived from a polyene, preferably vinylnorbornene or norbornadiene, is from 0.1 to 50 mol%, preferably from 0.1 to 20 mol%, and the proportion of the acyclic monoolefin of the formula VII is from 0 to 99.9 mol%, preferably from 5 to 80 mol%, based on the overall composition of the cycloolefin polymer. In the terpolymers described, the proportion of the polycyclic monoolefin is from 0.1 to 99.9 mol%, preferably from 3 to 75 mol%, based on the overall composition of the cycloolefin polymer.

Further amorphous polymers suitable for use according to the invention are described in EP-A-317262. Hydrogenated polymers and copolymers, e.g. of styrene or dicyclopentadiene, and other amorphous polyolefins are likewise suitable.

The cycloolefin copolymers used according to the invention can be prepared at temperatures of from -78 to 200°C at a pressure of from 0.01 to 200 bar in the presence of one or more catalyst systems comprising at least one transition metal compound and, if appropriate, a cocatalyst and, if appropriate, a support material. Suitable transition metal compounds include metallocenes, in particular stereorigid

metallocenes. Examples of catalyst systems which are suitable for the preparation of the cycloolefin copolymers used according to the invention are described in US-A-5,008,356, EP-A-0 407 870, EP-A-0 485 893 and EP-A-0 503 422. These references are hereby expressly incorporated by reference into the present patent application.

Examples of transition metal compounds used are:

rac-dimethylsilyl bis(1-indenyl)zirconium dichloride,
rac-dimethylgermyl bis(1-idenyl)zirconium dichloride,
rac-phenylmethlsilyl bis(1-indenyl)zirconium dichloride,
rac-phenylvinylsilyl bis(1-indenyl)zirconium dichloride,
1-silacyclobutyl bis(1-indenyl)zirconium dichloride,
rac-diphenylsilyl bis(1-indenyl)hafnium dichloride,
rac-phenylmethylsilyl bis(1-indenyl)hafnium dichloride,
rac-diphenylsilyl bis(1-indenyl)zirconium dichloride,
rac-ethylene-1,2 bis(1-indenyl)-zirconium dichloride,
dimethylsilyl(9-fluorenyl)(cyclopentadienyl)zirconium dichloride,
diphenylsilyl(9-fluorenyl)(cyclopentadienyl)zirconium dichloride,
bis(1-indenyl)zirconium dichloride,
diphenylmethylene(9-fluorenyl)cyclopentadienylzirconium dichloride,
isopropylene(9-fluorenyl)cyclopentadienylzirconium dichloride,
rac-isopropylidene bis(1-indenyl)zirconium dichloride,
phenylmethylmethylene(9-fluorenyl)cyclopentadienylzirconium dichloride,
isopropylene(9-fluorenyl) (1-(3-isopropyl)cyclopentadienyl)zirconium dichloride,
isopropylene(9-fluorenyl)(1-(3-methyl)cyclopentadienyl)zirconium dichloride,
diphenylmethylene(9-fluorenyl)(1-(3-methyl)cyclopentadienyl)zirconium dichloride,

methylphenylmethylene(9-fluorenyl)(1-(3-methyl)cyclopentadienyl)zirconium dichloride, dimethylsilyl(9-fluorenyl)(1-(3-methyl)cyclopentadienyl)zirconium dichloride, diphenylsilyl(9-fluorenyl)(1-(3-methyl)cyclopentadienyl)zirconium dichloride, diphenylmethylene(9-fluorenyl)(1-(3-tert-butyl)cyclopentadienyl)zirconium dichloride, isopropylene(9-fluorenyl)(1-(3-tert-butyl)cyclopentadienyl)zirconium dichloride, isopropylene(cyclopentadienyl)(1-indenyl)zirconium dichloride, diphenylcarbonyl(cyclopentadienyl)(1-indenyl)zirconium dichloride, dimethylsilyl(cyclopentadienyl)(1-indenyl)zirconium dichloride, isopropylene(methylcyclopentadienyl)(1-indenyl)zirconium dichloride, 4-(η^5-cyclopentadienyl)-4,7,7-trimethyl(η^5-4,5,6,7-tetrahydroindenylzirconium dichloride, [4-(η^5-cyclopentadienyl)-4,7,7-triphenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-cyclopentadienyl)-4,7-dimethyl-7-phenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'-tert-butylcyclopentadienyl)-4,7,7-triphenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'-tert-butylcyclopentadienyl)-4,7-dimethyl-7-phenyl-(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'methylcyclopentadienyl)-4,7,7-trimethyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'-methylcyclopentadienyl)-4,7,7-triphenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'-methylcyclopentadienyl)-4,7-dimethyl-7-phenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride, [4-(η^5-3'isopropylcyclopentadienyl)-4,7,7-trimethyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride,

[4-(η^5-3'isopropylcyclopentadienyl)-4,7,7-triphenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride,
[4-(η^5-3'isopropylcyclopentadienyl)-4,7-dimethyl-7-phenyl(η^5-4,5,6,7-tetrahydroindenyl)]zirconium dichloride,
[4-(η^5-cyclopentadienyl)(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-cyclopentadienyl)-4-methyl(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-cyclopentadienyl)-4-phenyl(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-cyclopentadienyl)-4-phenyl(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-3'-methylcyclopentadienyl)(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-3'-isopropylcyclpentadienyl)(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[4-(η^5-3'-benzylcyclopentadienyl)(η^5-4,5-tetrahyropentalene)]zirconium dichloride,
[2,2,4-trimethyl-4-(η^5-cyclopentadienyl)(η^5-4,5-tetrahydropentalene)]zirconium dichloride,
[2,2,4-trimethyl-4-(η^5-(3,4-diisopropyl)cyclopentadienyl)(η^5-4,5-tetrahydropentalene)]zirconium dichloride.

The cycloolefin copolymers can also be prepared in other ways which will be briefly outlined in the following: catalyst systems based on mixed catalysts comprising titanium salts and organoaluminum compounds are described in DD-A-109 224 and DD-A-237 070. EP-A-0 156 464 describes the preparation of cycloolefin copolymers due to catalysts based on vanadium.

The cycloolefin copolymers can also be prepared by ring-opening polymerization of at least one of the monomers of the formulae I to VI and subsequent hydrogenation of the products obtained.

The polymerization can also be carried out in a plurality of stages, in which block copolymers can also be formed (DE-A-42 05 416).

Cycloolefin copolymers are preferably amorphous, transparent materials. The heat distortion resistance of the cycloolefin copolymers can be set within a wide range. The glass transition temperature measured in accordance with DIN EN ISO 11357-1 can be employed as an indication of the heat distortion resistance as can be determined on injection-molded specimens in accordance with ISO 75 part 1 and part 2. The cycloolefin copolymers described have glass transition temperatures of from -50 to 220°C. Preference is given to glass transition temperatures of from 0 to 180°C, and particular preference is given to glass transition temperatures of from 40 to 180°C.

The mean molar mass of the cycloolefin copolymers can be controlled in a known manner by introduction of hydrogen, variation of the catalyst concentration or variation of the temperature. The cycloolefin copolymers to be used according to the invention have mass average molar masses Mw of from 1000 to 10,000,000 g/ml. Preference is given to mass average molar masses M_w of from 5000 to 5,000,000 g/mol, and particular preference is given to mass average molar masses M_w of from 5000 to 1,200,000 g/mol. These molar masses determined by means of gel permeation chromatography (GPC) in chloroform at 35°C with the aid of an RI detector are relative and based on a calibration using narrow-distribution polystyrene standards.

The cycloolefin copolymers described here have viscosity numbers measured in accordance with DIN 53 728 of from 5 to 5000 ml/g. Preference is given to viscosity numbers of from 5 to 2000 ml/g, and particular preference is given to viscosity numbers of from 5 to 1000 ml/g.

The optical properties of the polymer mixtures were determined on 1 mm thick pressed plates using a Gardner Haze-gard and in accordance with ASTM D 1003.

The process of the invention for producing a bimodal or multimodal mixture of one or more amorphous polyolefins regardless of the difference in the mean molar mass and in the molar mass distribution of the amorphous polyolefins is preferably carried out according to one or more of the process variants 1 to 5.

Process variant 1 as shown in Figure 1 comprises an assembly of two ($z = 0$) or more ($z > 0$) reactors connected in parallel, where the amorphous polyolefin having a high molar mass of VN > 100 ml/g and M_w > 100,000 g/mol is prepared by solution polymerization in one reactor and the other constituents of the mixture are produced in the other reactor or reactors. The reaction in these does not necessarily have to be carried out in solution. The combined solution is subsequently homomgenized and the solvent is separated off.

Process variant 2 as shown in Figure 2 comprises an assembly of two ($z = 0$) or more ($z > 0$) reactors connected in series, where the amorphous polyolefin having a high molar mass of VN > 100 ml/g and M_w > 100,000 g/mol is prepared by solution polymerization in the first reactor and the other constituents of the mixture are produced in the subsequent reactors. Catalyst, cocatalyst and monomers can be metered in individually upstream of each reactor. The combined solution is subsequently homogenized and the solvent is separated off.

Process variant 3 as shown in Figure 3 preferably employs a reactor in which the amorphous polyolefin having a high molar mass of VN > 100 ml/g and M_w >

100,000 g/mol is prepared by solution polymerization. The other constituents of the mixture are metered into the outflowing solution as a polymer solution having a concentration of from 1 to 100% by weight in, preferably, a hydrocarbon or hydrocarbon mixture. In the case of a solution containing 100% by weight of polymer, a polymer melt is present. The combined solution is subsequently homogenized and the solvent is separated off.

Process variant 4 preferably employs a reactor in which the amorphous polyolefin having a high molar mass of VN > 100 ml/g and M_w > 100,000 g/mol and the other constituents of the mixture are prepared simultaneously by solution polymerization using a catalyst combination. The solvent is subsequently separated off.

Process variant 5 preferably employs a reactor in which the amorphous polyolefin having a high molar mass of VN > 100 ml/g and M_w > 100,000 g/mol and the other constituents of the mixture are prepared in succession by periodic variation over time of the regulated concentration, preferably hydrogen, propylene, α-olefins or aluminum alkyls in such a way that, regarded over the mean residence time, a constant molar mass distribution is obtained. The solvent is subsequently separated off.

Preference is given to the process variants 1 to 3.

In general, the mechanical and rheological properties of polymers are dependent on the molar mass. The higher the molecular weight, the higher the elasticity, stiffness, creep resistance, viscosity, melt viscosity, environmental stress cracking resistance, chemical resistance, etc.

However, the use of high molecular weight, amorphous polyolefins having a narrow molar mass distribution is

not possible for many applications because their processing properties, mainly due to the low flowability, are unsatisfactory. The use of amorphous polyolefins having a broad molar mass distribution offers the opportunity of combining the good mechanical and rheological properties of relatively high molecular weight polyolefins with the excellent processability of low molecular weight polymers.

The bimodal or multimodal mixture of the invention can be used, in particular, for producing films having a better environmental stress cracking resistance and a reduced gel content, optical storage media (CD, DVD) having increased flowability and good chemical resistance, bottles and containers having improved environmental stress cracking resistance and chemical resistance, plastic articles having improved sterilizability by means of hot steam, gamma rays or electron beams, toner binders having improved fixing properties, e.g. broader antioffset window and higher printing speed, films and injection-molded articles having higher elasticity and stiffness, coatings having increased environmental stress cracking resistance and chemical resistance and films having improved barrier properties.

The invention is illustrated by the examples below:

Example 1

An ethylene-norbornene copolymer having a VN of 220 ml/g, a mass average molar mass of 280,000 g/mol and a glass transition temperature of 70°C is prepared continuously as a 6% strength by weight solution in decalin using process variant 3. A 50% strength by weight solution of another ethylene-norbornene copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol and a glass transition temperature of 65°C in decalin is then metered continuously into

the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 8 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a transparency of 93.3% and a haze of 2.5%. This demonstrates the high homogeneity of the mixture.

Example 2

An ethylene-norbornene copolymer having a VN of 220 ml/g, a mass average molar mass of 280,000 g/mol and a glass transition temperature of 70°C is prepared continuously as a 6% strength by weight solution in decalin using process variant 3. A melt of another ethylene-norbornene copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol and a glass transition temperature of 65°C in Exxol is then metered continuously into the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 15 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a high transparency and low light scattering, indications of good homogeneity. A pressed plate of this product has a transparency of 93.0% and a haze of 2.8%. This demonstrates the high homogeneity of the mixture.

Example 3

An ethylene-norbornene copolymer having a VN of 130 ml/g, a mass average molar mass of 120,000 g/mol and a glass transition temperature of 85°C is prepared continuously as a 20% strength by weight solution in decalin using process variant 3. A 50% strength by weight solution of another ethylene-norbornene

copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol and a glass transition temperature of 65°C in decalin is then metered continuously into the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 25 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a transparency of 93.5% and a haze of 2.4%. This demonstrates the high homogeneity of the mixture.

Example 4

An ethylene-norbornene copolymer having a VN of 130 ml/g, a mass average molar mass of 120,000 g/mol and a glass transition temperature of 135°C is prepared continuously as a 10% strength by weight solution in decalin using process variant 3. A 20% strength by weight solution of another ethylene-norbornene copolymer having a VN of 55 ml/g, a mass average molar mass of 60,000 g/mol and a glass transition temperature of 135°C in decalin is then metered continuously into the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 10 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a transparency of 94.0% and a haze of 1.9%. This demonstrates the high homogeneity of the mixture.

Example 5

An ethylene-norbornene copolymer having a VN of 220 ml/g, a mass average molar mass of 280,000 g/mol and a glass transition temperature of 80°C is prepared continuously as a 6% strength by weight solution in decalin using process variant 3. A 50% strength by

weight solution of another amorphous polyolefin having a glass transition temperature of 65°C (trade name: Escorez 5320) in decalin is then metered continuously into the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 10 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a transparency of 92.9% and a haze of 3.0%. This demonstrates the high homogeneity of the mixture.

Example 6

An ethylene-norbornene copolymer having a VN of 80 ml/g, a mass average molar mass of 100,000 g/mol and a glass transition temperature of 80°C is prepared continuously as a 6% strength by weight solution in decalin using process variant 3. A 50% strength by weight solution of another amorphous polyolefin having a glass transition temperature of 65°C (trade name: Escorez 5320) in decalin is then metered continuously into the first ethylene-norbornene copolymer solution in such an amount that the proportion of the higher molecular weight polymer is 25 percent by weight of the total mass of polymer. The solution is mixed well and the solvent is removed thermally using known techniques. A pressed plate of this product has a transparency of 93.2% and a haze of 2.8%. This demonstrates the high homogeneity of the mixture.

Comparative Example 1

In a Haake TW 100 twin-screw extruder, an ethylene-norbornene copolymer having a VN of 80 ml/g, a mass average molar mass of 100,000 g/mol and a glass transition temperature of 75°C is mixed in the melt with another ethylene-norbornene copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol

and a glass transition temperature of 65°C in such an amount that the final mixture contains 10% of the higher molecular weight amorphous polyolefin. The extruded strand of melt can clearly be seen to contain non-melted droplets which scatter light more strongly. The transparency is 89.9% and the haze is 8.8%. This is an indication of the poor homogeneity of the mixture.

Comparative Example 2

In a Haake TW 100 twin-screw extruder, an ethylene-norbornene copolymer having a VN of 80 ml/g, a mass average molar mass of 100,000 g/mol and a glass transition temperature of 75°C is mixed in the melt with another ethylene-norbornene copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol and a glass transition temperature of 65°C in such an amount that the final mixture contains 20% of the higher molecular weight amorphous polyolefin. The transparency is 91.4% and the haze is 3.8%. This is an indication of the poor homogeneity of the mixture.

Comparative Example 3

In a Haake TW 100 twin-screw extruder, an ethylene-norbornene copolymer having a VN of 80 ml/g, a mass average molar mass of 100,000 g/mol and a glass transition temperature of 75°C is mixed in the melt with another ethylene-norbornene copolymer having a VN of 15 ml/g, a mass average molar mass of 12,000 g/mol and a glass transition temperature of 65°C in such an amount that the final mixture contains 50% of the higher molecular weight amorphous polyolefin. The transparency is 92.8% and the haze is 3.0%. This is an indication of satisfactory homogeneity of the mixture.

Comparative Example 4

An attempt is made to mix 85% by weight of an ethylene-norbornene copolymer having a VN of 20 ml/g and a glass transition temperature of 63°C with 5% by weight of an ethylene-norbornene copolymer having a VN of 81 ml/g and a glass transition temperature of 75°C and 10% by weight of an ethylene-norbornene copolymer having a VN of 115 ml/g and a glass transition temperature of 75°C by means of a Haake TW 100 twin-screw extruder. The extruded strand can clearly be seen to contain unmelted droplets which scatter light more strongly. The transparency is 87.3% and the haze is 9.8%. This is an indication of the satisfactory homogeneity of the mixture.

Comparative Example 5

An attempt is made to mix 80% by weight of an ethylene-norbornene copolymer having a VN of 20 ml/g and a glass transition temperature of 63°C with 5% by weight of an ethylene-norbornene copolymer having a VN of 81 ml/g and a glass transition temperature of 75°C and 15% by weight of an ethylene-norbornene copolymer having a VN of 115 ml/g and a glass transition temperature of 75°C by means of a Haake TW 100 twin-screw extruder. The extruded strand can clearly be seen to contain unmelted droplets which scatter light more strongly. The transparency is 88.7% and the haze is 5.8%. This is an indication of the poor homogeneity of the mixture.

Comparative Example 6

An attempt is made to mix 70% by weight of an ethylene-norbornene copolymer having a VN of 20 ml/g and a glass transition temperature of 63°C with 15% by weight of an ethylene-norbornene copolymer having a VN of 81 ml/g and a glass transition temperature of 75°C and 15% by weight of an ethylene-norbornene copolymer having a VN of 115 ml/g and a glass transition temperature of 75°C by means of a Haake TW 100 twin-screw extruder. The

extruded strand can clearly be seen to contain unmelted droplets which scatter light more strongly. The transparency is 90.7% and the haze is 5.3%. This is an indication of the poor homogeneity of the mixture.

Comparative Example 7

An attempt is made to extrude an ethylene-norbornene copolymer having a VN of 200 ml/g and a glass transition temperature of 65°C through a 1 × 1 mm nozzle at a temperature of 310°C and a shear stress of 2.7E5 Pa. The extruded extrudate of melt displays strong brown discoloration and has severe structuring of the surface. A viscosity of 100,000 Pas and an MFI of 0.27 cm^3/10 min are determined. At 280°C, no strand at all is obtained. Such a product can accordingly be processed in the melt only with extreme difficulty.

The difference in the materials properties, in particular the optical properties, between products prepared in the examples and in the comparative examples are shown in Table 1 below.

Table 1

Example (E) Comparative Example (C)	Transparency (%)	Haze (%)
E1	93.3	2.5
E2	93.0	2.8
E3	93.5	2.4
E4	94.0	1.9
E5	92.9	3.0
E6	93.2	2.8
C1	89.9	8.8
C2	91.4	3.8
C3	92.8	3.0
C4	87.3	9.8
C5	88.7	5.8
C6	90.7	5.3

Claims

1. A process for the continuous preparation of a bimodal or multimodal mixture of two or more amorphous polyolefins having a different molar mass, wherein the viscosity ratio of at least two amorphous polyolefins having a different molar mass is less than 0.005 and greater than 4 and a) the amorphous polyolefin having a high molar mass is prepared by solution polymerization in one reactor of an assembly of two or more reactors connected in parallel or in series and the other constituents of the mixture are produced in the other reactors after which the polyolefins are mixed in solution, or b) the amorphous polyolefin having a high molar mass is prepared by solution polymerization in one reactor and the other constituents of the mixture are introduced in the form of a polymer solution into the solution flowing from the reactor, and the solution of polymer mixture obtained according to a) or b) is homogenized and the solvent is separated off.

2. The process as claimed in claim 1, wherein the amorphous polyolefin having a high molar mass has a VN of > 80 ml/g and an M_w of > 90,000 g/mol.

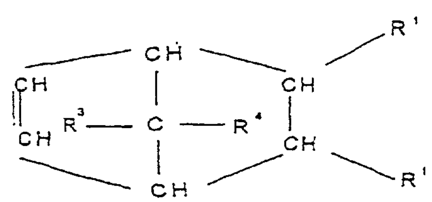
3. The process as claimed in claim 1, wherein the amorphous polyolefin having a high molar mass has a VN of > 100 ml/g and an M_w of > 100,000 g/mol.

4. The process as claimed in claim 1, wherein the amorphous polyolefin having a high molar mass has a VN of > 120 ml/g and an M_w of > 120,000 g/mol.

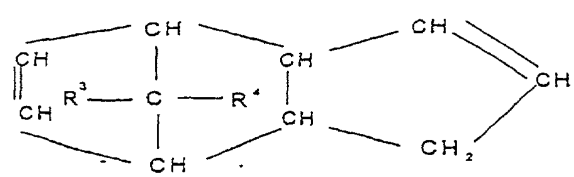
5. The process as claimed in claim 1, wherein the amorphous polyolefin having a high molar mass has a VN of > 150 ml/g and an M_w of > 150,000 g/mol.

6. The process as claimed in any of claims 1 to 5, wherein the amorphous polyolefin is a cycloolefin copolymer.

7. The process as claimed in one or more of claims 1 to 6, wherein the bimodal or multimodal mixture comprises at least one cycloolefin copolymer comprising from 0.1 to 100% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from at least one polycyclic olefin of the formula I, II, II', III, IV, V or VI.



(I),



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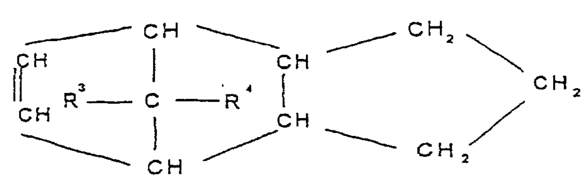

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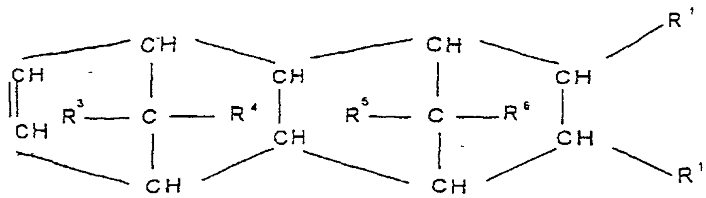
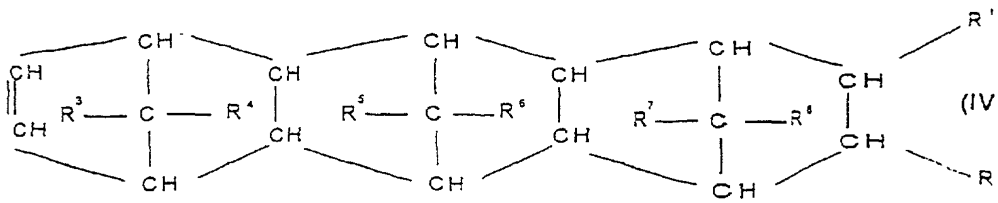


(III),



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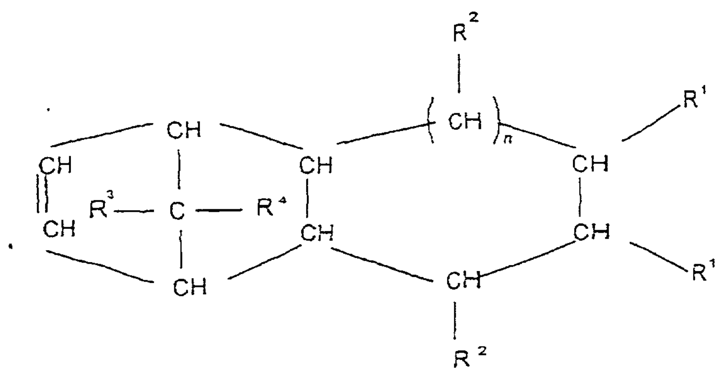
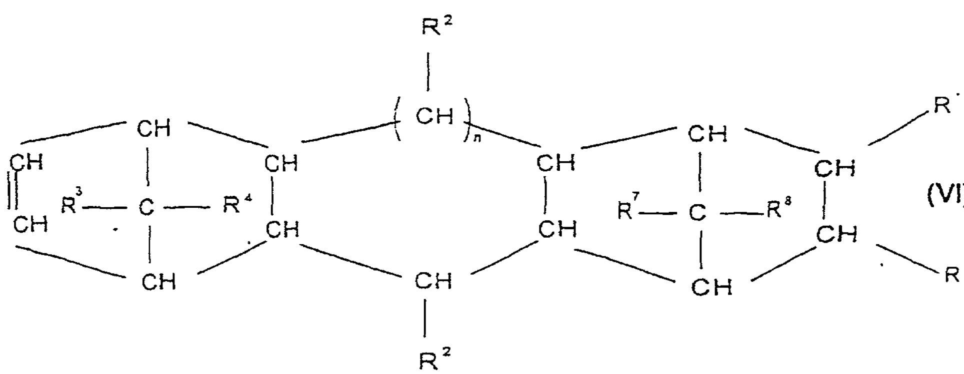


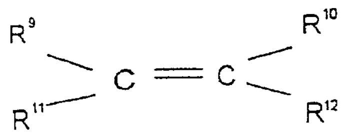
(V),



(VI),

where R^1, R^2, R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and are each a hydrogen atom or a C_1-C_{20}-hydrocarbon radical such as a linear or branched C_1-C_8-alkyl radical, a C_6-C_{18}-aryl radical, a C_7-C_{20}-alkylenearyl radical or a cyclic or acyclic C_2-C_{20}-alkenyl radical or form a saturated, unsaturated or

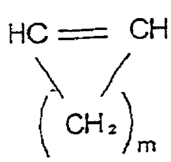
aromatic ring, where identical radicals R^1 to R^8 in the various formulae I to VI can have different meanings, and n can be from 0 to 5, and, if desired, up to 99.9% by weight, based on the total mass of the cycloolefin copolymer, of polymerized units derived from one or more acyclic olefins of the formula VII



(VII),

where R^9, R^{10}, R^{11} and R^{12} are identical or different and are each a hydrogen atom, a linear, branched, saturated or unsaturated C_1-C_{20}-hydrocarbon radical such as a C_1-C_8-alkyl radical or a C_6-C_{18}-aryl radical.

8. The process as claimed in one or more of claims 1 to 7, wherein the cycloolefin copolymers further comprise up to 45% by weight, based on the total mass of the cycloolefin copolymer, or polymerized units derived from one or more monocyclic olefins of the formula VIII



(VIII),

where m is from 2 to 10.

9. The process as claimed in one or more of claims 1 to 8, wherein the cyclic and polycyclic olefins contain one or more of the groups halogen, hydroxyl, ester, alkoxy, carboxy, cyano, amido, imido and silyl.

10. The process as claimed in one or more of claims 1 to 9, wherein the cycloolefin copolymers comprise

polymerized units derived from polycyclic olefins of the formula I or III and polymerized units derived from acyclic olefins of the formula VII.

11. The process as claimed in one or more of claims 1 to 10, wherein the cycloolefin copolymers comprise polymerized units derived from olefins having a norbornene skeleton, preferably from norbornene, tetracyclododecene, vinylnorbornene or norbornadiene.

12. The process as claimed in one or more of claims 1 to 11, wherein the cycloolefin copolymers comprise polymerized units derived from acyclic α-olefins having from 2 to 20 carbon atoms, preferably ethylene or propylene, particularly preferably ethylene.

Figure 1

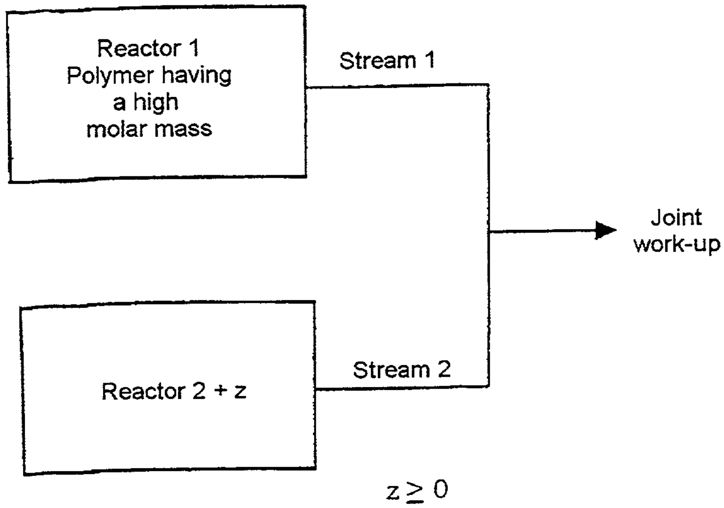

Reactor 1
Polymer having
a high
molar mass
Stream 1
Joint
work-up
Reactor 2 + z
Stream 2
z ≥ 0

Figure 1

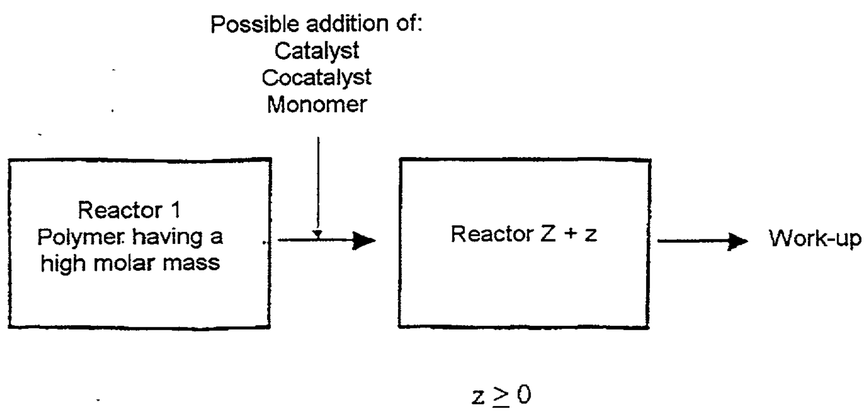
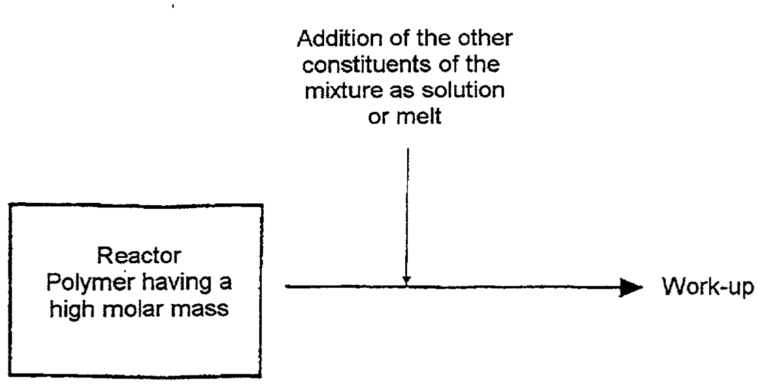


Figure 3



ATTORNEY DOCKET NO.: 1999/G 014 (5587*324) #4

JC10 Rec'd PCT/PTO 27 DEC 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

10/018,847

APPLICANT: FRANK OSAN *ET AL.*	
INTERNATIONAL APPL. NO.: PCT/EP00/05759	
SERIAL NO. TO BE ASSIGNED	ART UNIT: TO BE ASSIGNED
FILED:	EXAMINER: TO BE ASSIGNED
FOR: METHOD OF PRODUCING AMORPHOUS POLYOLEFINS WITH A WIDE MOLE WEIGHT DISTRIBUTION	

place fees und EP #. priority 1/9/02

Asst. Commissioner for Patents
Washington, D.C. 20231

"EXPRESS MAIL" No. EL928735645US DATE: December 27, 2001

I HEREBY CERTIFY THAT THIS PAPER OR FEE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE "EXPRESS MAIL POST OFFICE TO ADDRESSEE" SERVICE UNDER 37 CFR 1.10 ON THE DATE INDICATED AND IS ADDRESSED TO THE U.S. PATENT AND TRADEMARK OFFICE, P.O. BOX 2327, ARLINGTON, VA 22202

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(TYPED OR PRINTED NAME OF

Carrie A. McPherson
(SIGNATURE OF PERSON MAILING

10018847 .122701

TRANSMITTAL OF MISSING PARTS

Sir:

Please find the enclosed papers:

1) Original Declaration/Power of Attorney form executed by inventors:
2) The undersigned's check for $130.00 to cover the surcharge for missing parts.
3) A copy of the Notice to File Missing Parts has not yet been received.

The Commissioner is authorized to debit any deficiency, or credit any overpayment, of the above-mentioned fees to our Deposit Account No. 03-2775.

Respectfully submitted,

CONNOLLY, BOVE, LODGE & HUTZ, LLP

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01/09/2002 UEDUVIJE 00000099 PCT/EP00/05759
01 FC:154 130.00 OP

Adjustment date: 03/20/2002 AYILMAZ
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01 FC:154 -130.00 OP

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#4

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:
My residence, post office address and citizenship are as stated below next to my name.
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Process for producing amorphous polyolefins having a broad molar massdistribution

the specification of which

(check one) is attached hereto.

Application.Serial.No. ______________________ and

was.amended.on ______________________ (if applicable)

was amended through ______________________

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s) for which Priority is Claimed:

Federal Republic of Germany, 19929809.2 of June 30, 1999

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States application(s) listed below.

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

(Application Serial No.)	(Filing Date)	(Status)
PCT/EP00/05759	21. June 2000	Pub. 01/02481

1999/G014 US PCT

(Application Serial No.) (Filing Date) (Status)
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

In the matter of the above-identified application, please recognize Rudolf E. Hutz, Reg. No. 22,397; John D. Fairchild, Reg. No. 19,756; Harold Pezzner, Reg. No. 22,112; Richard M. Beck, Reg. No. 22,580; Paul E. Crawford, Reg. No. 24,397; Patricia Smink Rogowski, Reg. No. 33,791; Robert G. McMorrow, Jr., Reg. No. 30,962; Ashley I. Pezzner, Reg. No. 35 646, William E. McShane, Reg. No. 32,707; Mary W. Bourke, Reg. No. 30,982; Gerard M O'Rourke, Reg. No. 39,794; James M. Olsen, Reg. No. 40,408; Francis DiGiovanni, Reg. No. 37,310; Eric J. Evain, Reg. No. 42,517; Daniel C. Mulveny, Reg. No. 45,897, Christine M. Hansen, Reg. No. 40,634; Patrick H. Higgins 39,709 and Elliot C. Mendelson (Agent), Reg. No. 42,878, all of P.O. Box 2207, Wilmington, Delaware 19899-2207 as attorneys with full power of substitution to prosecute this application and conduct all business in the Patent and Trademark Office connected therewith.

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